ECE LABORATORY IN THE VINČA INSTITUTE – ITS BASIC CHARACTERISTICS AND FUNDAMENTALS OF ELECTROCHEMICAL ETCHING ON POLYCARBONATE

by

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This paper deals with the introductory aspects of the Electrochemical Etching Laboratory installed at the VINČA Institute in the year 2003. The main purpose of the laboratory is its field application for radon and thoron large-scale survey using passive radon/thoron UFO type detectors. Since the etching techniques together with the laboratory equipment were transferred from the National Institute of Radiological Sciences, Chiba, Japan, it was necessary for both etching conditions to be confirmed and to be checked up, *i. e.*, bulk etching speeds of chemical etching and electrochemical etching in the VINČA Electrochemical Etching Laboratory itself. Beside this initial step, other concerns were taken into consideration in this preliminary experimental phase such as the following: the measurable energy range of the polycarbonate film, background etch pit density of the film and its standard deviation and reproducibility of the response to alpha particles for different sets of etchings.

Key words: electrochemical etching, radon, thoron, polycarbonate film, bulk etching rate

INTRODUCTION

In the period from 1998 to 2000 indoor radon and thoron measurements were carried out by VINČA Institute in Gornja Stubla field – in Kosovo and Metohia Province [1-5] by passive radon UFO type detectors designed and produced in 1992 at the National Institute for Radiological Sciences (NIRS), Chiba, Japan [6]. The results obtained showed there were both high radon and high thoron levels in Yugoslavia [7].

Under the research agreement on cooperation between NIRS and VINČA Institute signed in 2002, the laboratory equipment was brought by NIRS in the same year, and in the year 2003 the Laboratory for electrochemical etching (ECE Laboratory) was founded after the renovation of an old laboratory at the VINČA Institute. Since the proposed etching condition for radon/thoron discriminative passive UFO type detectors requires room temperature to be maintained at 30 degrees Celsius, the walls and window of the room were reconstructed in order to have the necessary high energy insulation capability. New electric power lines were installed to provide sufficient power supply for the equipment. The sink was also renewed to be able to handle etching cell units used for the chemical and electrochemical etching.

After the transfer of the ECE Laboratory from NIRS to VINČA Institute it was necessary to confirm reliability of the laboratory in the new conditions. The characteristics of the electrochemical etching procedure used is an essential in the process of enlargement of latent alpha particles tracks. The complete experimental procedure performed in NIRS in Japan [8] was repeated in the new environment at VINČA Institute with a few new additional features. It should be noted that the process of electrochemical etch pit formation is still not completely understood [9].

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BASIC CHARACTER OF THE ELECTROCHEMICAL ETCHING ON POLYCARBONATE

The main goal of the laboratory is to carry out radon/thoron measurements, for this purpose the UFO type of passive detector was developed. This UFO type detector, fig. 1, which was used in Japanese national program of radon survey [10, 11], consists of two hemispheres. One polycarbonate film is placed in each hemisphere. In the upper, larger thoron and radon (Tn & Rn) hemisphere, the film detects both radon and thoron with their progenies. The smaller, radon (Rn) hemisphere is connected to the larger hemisphere through a single pinhole with diameter of 1 mm. Diffusion to the Rn hemisphere is small enough to practically prevent thoron from entering the hemisphere, because of its short half-life of 55 seconds. Radon with longer half-life of 3.8 days easily enters the Rn hemisphere. Therefore, the film in the Rn hemisphere detects only radon and its daughters.

The alpha particles originating from thoron alpha decay have higher energy than the ones originating from radon alpha decay. The films from Rn hemisphere are etched for 0.5 h of chemical pre-etching and 3 h of electrochemical etching (resulting energy window 0.85-2.7 MeV). The films from Tn & Rn hemisphere are etched for 3 h of chemical etching (CE) and 3 h of electrochemical etching (resulting energy window 2.5-4.0 MeV) [12].

Iupilon polycarbonate films (Mitsubishi Gas Chemical Company, Inc., Japan) were used in the experiments. The films are round in shape with a diameter of 50 mm and a thickness of 300 μm. The sensitive side of the film is covered with a 50 μm thick foil. The purpose of the foil is both to protect the film from mechanical damage and alpha radioactivity in the environment. The chemical form of polycarbonateis–$[\text{-O-}C_6H_4C\text{-}(CH_3)_2C_6H_4OCO\text{-}]_n$-. The etching solution used in the experiment was 8N KOH (80% volume fraction) and C_2H_5OH (20% volume fraction). The condition for etching of polycarbonate films for radon and thoron measurements is room temperature of 30 °C. A solution of 1N KOH is used only as a conductor on the other side of the films, allowing application of high electrical fields on the polycarbonate films.

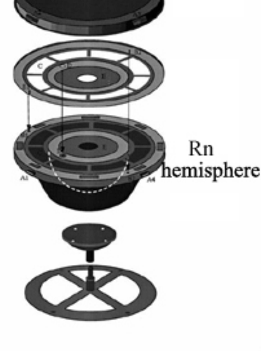

Figure 1. Discriminative radon passiv UFO type detector

The power supply for etching cells is 800 V and 2 kHz, and it is used during the electrochemical etching period. Beside the power supply unit, there is a control unit whose major purpose is to shut down the power supply only to a damaged etching cell in case of a short-out. The short-out occurs for two major reasons; complete dielectric breakthrough of polycarbonate film and because of the leakage of solution from cells. In this case the control unit cuts the power supply only in these cells and in other cells etching continues undisturbed. A circular shaped area of film with diameter of 32 mm is etched.

EXPERIMENTAL EXERCISE

A mass measurement method was used for the estimation of bulk etching rate. The mass of two sets of 20 films was measured, before and after the chemical and electrochemical treatment. Bulk etching rate was evaluated from the weight difference. In order to estimate temperature dependence of bulk etching rate, measurements were carried out at temperatures of 20, 25, and 30 °C. Prior to aplanned etching, all equipment and solutions were kept overnight at a specified temperature, as one of the standard procedures transferred from NIRS. All these precautions were taken in order to ensure that the specified temperature is achieved, because chemical and electrochemical bulk etching rates are mostly temperature dependent. The ethanol fraction dependence of bulk etching rate was estimated at 20, 40, and 60% of the volume fraction of ethanol in the etching solution.

For estimation of the bulk etching rate of chemical etching, films were etched for between 4 and 5 hours. Pouring etchant into the cells takes 2 minutes. It was considered that the middle of this period (1 minute after the initiation) was to be the start time of the etching process. A very important matter in this method of measuring chemical bulk etching rate was manual shaking of the etching cells for at least 10 minutes because air bubbles might be formed during the pouring of the etchant into the cells. The same method was applied for the estimation of the electrochemical bulk etching rate as well. The electrochemical etching process was performed after 15 minutes of chemical etching. These 15 minutes of the chemical etching are unavoidable, because the pouring of the etchant into cells and the shaking of cells was time consuming, and they were not performed during the electrochemical etching. After 15 minutes of chemical etching, films were electrochemically etched for around 4 hours. The electrochemical bulk etching rate was then estimated from the weight difference taking into account the 15 minutes of chemical etching.

Two standard sources of Am-241 were used for irradiation of films with the activity of 2321 ± 20 Bq and 1400 ± 20 Bq respectively. For the purpose of energy window estimation, various distances between the source and film were chosen in order to achieve different energies of incident alpha particles. Fluctuation of incident alpha energy was between 2.5 and 5%.

RESULTS

The weight difference of the sets of 20 films, before and after the etching was from 200 mg to 400 mg, depending on the condition of etching (temperature, ethanol fraction). The obtained bulk etching rates for electrochemical and chemical etching were 4.07 μm/h and 4.26 μm/h, respectively at temperature of 30 °C and with solution 8N KOH and 20% of ethanol. The chemical bulk etching rate was 14% higher than that estimated in NIRS.

Higher bulk etching rate resulted in shifting the energy window, comparing it with the one estimate in NIRS for radon and thoron measurement by the UFO detector. The nominal energy windows estimate in NIRS were: 0.8-2.7 MeV, for etching of radon films (CE: 0.5 h + ECE: 3 h) and 2.5-4.0 MeV, for etching of thoron films (CE: 3 h + ECE: 3 h). The energy windows estimated in VINČA Institute were: for etching of radon films 0.75-3.5 MeV and for etching of thoron films 3.0-4.6 MeV. Variation of the reproducibility for energies in the middle of the energy window was less than 3%, and for energies at the borders of the energy window was 20-40%. Contribution of border energies to the sensitivity of the film for the expected whole energy window was 10%. Consequently the overall variation of reproducibility was estimated to be around 6%.

Background etch pit density for radon film etching (CE: 0.5 h +ECE: 3 h) was 3.9±2.4 tracks/cm^2. For the thoron film (CE: 3 h + ECE: 3 h), background etch pit density was 4.0±2.4 tracks/cm^2. For an etching with 0.25 h CE and then 3.9 h ECE, background etch pit density was 8.6±2.9 tracks/cm^2. This leads to a conclusion that 15 minutes of chemical pre-etching is not enough to erase mechanical damages of the surface of the polycarbonate films.

A linear ethanol fraction dependence of the bulk etching rate in the range from 20% to 60% of ethanol fraction was observed, fig. 2. The slope was 0.056 μm/h per one percent of fraction of ethanol. The linearity does not exist outside of this range – the bulk etching rate drops to zero for 0% and 100% of ethanol fraction. It was also observed that the linear temperature dependence of bulk etching rate (fig. 3) has the slope of 0.236 μm/h°C.

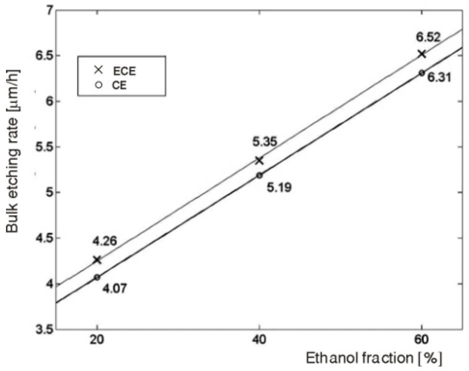

Figure 2. Ethanol fraction dependence of the bulk etching rate

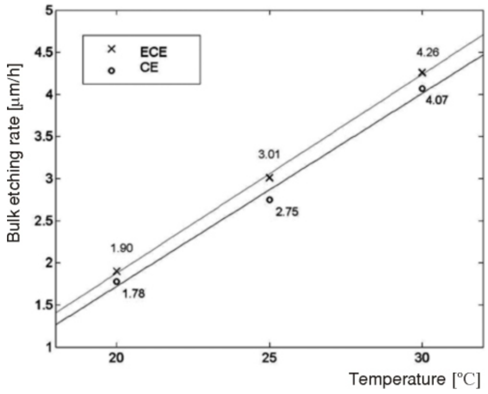

Figure 3. Temperature dependence of the bulk etching rate

CONCLUSION

The transfer of the ECE technology from Japan (NIRS) to Serbia and Montenegro (VINČA Institute) was completed successfully. Despite the fact that all the known conditions are the same, there might exist a difference in the basic characteristics of electrochemical etching when the transfer of technology was carried out. The present calibration factors might be changed from the previous ones. Therefore, it is necessary to conduct calibration exercises for our present etching conditions.

The VINČA ECE Laboratory has the potential to be a regional center for radon/thoron population exposure investigation in the Balkans but also for scientific training regarding future usage of the electrochemical etching devices. In addition, a nationwide survey of indoor radon/thoron concentrations in Serbia is planned with special attention being devoted to high natural radiation environmental regions and also to exposures in schools and kindergartens.

The ECE Laboratory can also be used in other applications, such as neutron flux measurements, neutron energy spectrum measurements, polymer lattice defect mechanism investigation, and development of new methods for radiation measurements.

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Зора С. ЖУНИЋ, Предраг УЈИЋ, Игор ЧЕЛИКОВИЋ, Кензо ФУЂИМОТО

ЛАБОРАТОРИЈА ЗА ЕЛЕКТРОХЕМИЈСКО РАЗВИЈАЊЕ ЧВРСТИХ НУКЛЕАРНИХ ТРАГ ДЕТЕКТОРА У ИНСТИТУТУ »ВИНЧА« – ОСНОВНЕ КАРАКТЕРИСТИКЕ И ПРИМЕНА

У раду се описује методологија мерења концентрације радона, торона и радонових потомака у ваздуху затворених просторија која се спроводи у новој Лабораторији за електрохемијско развијање нуклеарних траг детектора у Институту „Винча” (ЕЦЕ лабораторија). Опрема за лабораторију и методологија мерења представљају у потпуности трансфер технологије Националног института за радиолошке науке (National Institute of Radiological Sciences), Чиба, Јапан. Експериментално су проверене основне карактеристике електрохемијског развијања поликарбонатних филмова у новим лабораторијским условима у Институту „Винча”, и то: брзина нагризања површине поликарбонатних филмова хемијским и електрохемијским путем за номиналне услове, брзина нагризања филмова у зависности од температуре и у зависности од фракције етанола у раствору базе, зависност ширине и позиције енергетског прозора за детекцију алфа честица на поликарбонатном филму при номиналним трајањима хемијског и електрохемијског развијања, густина фона и поновљивост одзива поликарбонатног филма на алфа честице. Укратко су описани инструменти и опрема ЕЦЕ лабораторије, поликарбонатни филмови који служе као чврсти нуклеарни траг детектори за пасивни тип УФО радонског детектора и могућности примене електрохемијске методе и саме лабораторије у основном и примењеном истраживању.